

(REV 10/01)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

VER-154XX

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

10/009622

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 36 U.S.C. 371

INTERNATIONAL APPLICATION NO.

PCT/NL00/00404 ✓

INTERNATIONAL FILING DATE

13 June 2000 ✓ (13.06.00)

PRIORITY DATE CLAIMED

11 June 1999 ✓ (11.06.99)

TITLE OF INVENTION

METHOD FOR REMOVING NITROGEN OXIDES FROM AN OXYGEN-CONTAINING GAS STREAM

APPLICANT(S) FOR DO/EO/US

Willem Hendrik Bouwman, Petrus Franciscus Maria Theresia van Nisselrooij, Renee van Yperen, Sytse Jelle Jelles

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2)) in English
 - a. ☒ is attached hereto (required only if not transmitted by the International Bureau). Published appln WO 00/76637 A1
 - b. ☐ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
- ☐ An English language translation of the International Application as filed (35 U.S.C. 371(c)(3)).
 - a. ☐ is attached hereto.
 - b. ☐ had been previously submitted under 35 U.S.C. 154(d)(4).
- ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are attached herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
- ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
- ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
- ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 20. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:
 AMENDED CLAIMS PER INTERNATIONAL PRELIMINARY EXAMINATION REPORT DATED
 31 JULY 2001 (2 pages / issued in English)

U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <div style="font-size: 2em; font-weight: bold;">10/009622</div>		INTERNATIONAL APPLICATION NO PCT/NL00/00404		ATTORNEY'S DOCKET NUMBER VER-154XX	
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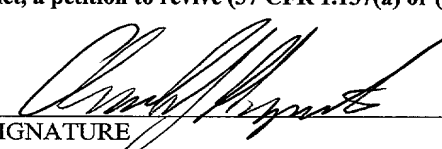
21. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1,040.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 <div style="text-align: right;">ENTER APPROPRIATE BASIC FEE AMOUNT =</div>				CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 0	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	23 - 20 =	3	X \$18.00	\$ 54.00	
Independent claims	1 - 3 =	0	X \$84.00	\$ 0	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ \$280.00	\$ 0
TOTAL OF ABOVE CALCULATIONS =				\$ 944.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$ --	
SUBTOTAL =				\$ 944.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$ 0	
TOTAL NATIONAL FEE =				\$ 944.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$ 0	
TOTAL FEES ENCLOSED =				\$ 944.00	
				Amount to be refunded:	\$
				charged:	\$

a.	<input checked="" type="checkbox"/>	A check in the amount of \$ <u>944.00</u> to cover the above fees is enclosed. A check in the amount of \$ _____ is enclosed for the assignment recordation fee.
b.	<input type="checkbox"/>	Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.
c.	<input checked="" type="checkbox"/>	The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>23-0804</u> . A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

Customer Number 207
 SEND ALL CORRESPONDENCE TO:

 Weingarten, Schurgin, Gagnebin & Lebovici LLP
 Ten Post Office Square
 Boston, Massachusetts 02109



SIGNATURE

NAME: Charles L. Gagnebin III
 REGISTRATION NUMBER: 25,467

Date: 12-11-1

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application : WILLEM HENDRIK BOUWMAN, ET AL.
Application No. :
Filed : Herewith
For : METHOD FOR REMOVING NITROGEN OXIDES FROM
AN OXYGEN-CONTAINING GAS STREAM
Examiner :
Attorney's Docket : VER-154XX

Group Art Unit:

I hereby certify that this correspondence is being deposited
with the United States Postal Service as first class mail in an
envelope addressed to: Commissioner for Patents, Washington,
D.C. 20231 on _____.

By: _____
Charles L. Gagnebin III
Registration No. 25,467
Attorney for Applicant(s)

PRELIMINARY AMENDMENT

BOX PCT
Commissioner for Patents
Washington, D.C. 20231

Sir:

Submitted for examination are the claims that were amended
by the International Preliminary Examination Report dated
July 31, 2001 (copy enclosed).

Kindly enter the following Preliminary Amendment in the
above-identified application:

Express Mail Number

EV 009950688 US

In the Claims:

In the Amended Claims of the International Preliminary Examination Report dated July 31, 2001 (copy enclosed):

Please amend the Claims to read as follows (a copy of the amended claims showing the additions and deletions appears at the end for the Examiner's convenience):

1. A method for reducing the content of nitrogen oxides in a nitrogen oxide- and oxygen-containing effluent by contacting this effluent in the presence of a deNOx catalyst, optionally in a NOx-storage system, with a reducing gas stream c, which comprises CO, H₂ and possibly NH₃ and which stream c has been obtained by converting a stream d, which comprises one or more hydrocarbons which are at least partly drawn from a different source than said effluent, with a stream e comprising oxygen and/or water from said effluent stream, if desired in the presence of a reductant forming catalyst, wherein substantially all of the oxygen present in stream e that is used for the formation of stream c is used up.

2. A method according to claim 1, wherein said effluent is the effluent of a step for combusting fuels, which comprises the steps of feeding at least a stream a comprising one or more

fuels and a stream b containing excess oxygen with respect to the fuel; wherein said effluent, together with said reducing gas stream c which further optionally comprises one or more hydrocarbons, is contacted with said deNOx catalyst; wherein to stream c no ammonia and no urea is added.

3. A method according to claim 1, wherein said effluent originates from a combined heat and power unit.

4. A method according to claim 1, wherein said stream c is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst.

5. A method according to claim 2, wherein the fuel in stream a comprises synthesis gas and/or one or more hydrocarbons.

6. A method according to claim 2, wherein stream a and stream d comprise at least partly the same compounds.

7. A method according to claim 2, wherein the fuel in stream a comprises hydrocarbons which, like the hydrocarbons in stream d, have been selected independently from the group consisting of

natural gas, methane, diesel oil, gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, and mixtures thereof.

8. A method according to claim 1, wherein a reductant forming catalyst is used which is a partial oxidation catalyst for the partial oxidation of hydrocarbons, selected from the group consisting of Pt, Rh, Ru, Pd, Co and Ni and combinations thereof, optionally provided on a support, which support is optionally stabilized with Si, La, Ba and/or Y.

9. A method according to claim 1, wherein the reductant forming catalyst is a steam reforming catalyst which comprises Ni, Rh and/or Pt.

10. A method according to claim 1, wherein the reductant forming step is a partial oxidation step, wherein the ratio of hydrocarbons and oxygen is selected such that no complete oxidation takes place.

11. A method according to claim 1, wherein nitrogen oxides from said effluent are absorbed onto a suitable absorbent and

subsequently said reducing gas stream is contacted with said absorbent.

13. A method according to claim 11, wherein the product that is formed in the step wherein stream c is contacted with the structured support is used as fuel for a gas engine.

Please add the following new claims 14-23:

14. A method according to claim 2, wherein said effluent originates from a combined heat and power unit.

15. A method according to claim 2, wherein said stream c is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst.

16. A method according to claim 3, wherein said stream c is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst.

17. A method according to claim 3, wherein the fuel in stream a comprises synthesis gas and/or one or more hydrocarbons.

18. A method according to claim 4, wherein the fuel in stream a comprises synthesis gas and/or one or more hydrocarbons.

19. A method according to claim 3, wherein stream a and stream d comprise at least partly the same compounds.

20. A method according to claim 4, wherein stream a and stream d comprise at least partly the same compounds.

21. A method according to claim 5, wherein stream a and stream d comprise at least partly the same compounds.

22. A method according to claim 12, wherein the product that is formed in the step wherein stream c is contacted with the structured support is used as fuel for a gas engine.

23. A method according to claim 21, wherein:

the fuel in stream a comprises hydrocarbons which, like the hydrocarbons in stream d, have been selected independently from the group consisting of natural gas, methane, diesel oil,

gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, and mixtures thereof;

a reductant forming catalyst is used which is a partial oxidation catalyst for the partial oxidation of hydrocarbons, selected from the group consisting of Pt, Rh, Ru, Pd, Co and Ni and combinations thereof, optionally provided on a support, which support is optionally stabilized with Si, La, Ba and/or Y;

the reductant forming catalyst is a steam reforming catalyst which comprises Ni, Rh and/or Pt;

the reductant forming step is a partial oxidation step, wherein the ratio of hydrocarbons and oxygen is selected such that no complete oxidation takes place;

nitrogen oxides from said effluent are absorbed onto a suitable absorbent and subsequently said reducing gas stream is contacted with said absorbent;

said absorbent is a barium and/or zeolite comprising alumina washcoat on a structured support;

the product that is formed in the step wherein stream c is contacted with the structured support is used as fuel for a gas engine.


REMARKS

This Preliminary Amendment puts the claims into proper form for examination. Note that claims 1-11 and 13 have been amended; new claims 14-23 have been added; and claim 12 remains unchanged. Kindly calculate the filing fee based on the amended claims.

The Examiner is encouraged to telephone the undersigned attorney to discuss any matter which would expedite allowance of the present application.

Respectfully submitted,

WILLEM HENDRIK BOUWMAN, ET AL.

By: 
Charles L. Gagnebin III
Registration No. 25,467
Attorney for Applicant(s)

WEINGARTEN, SCHURGIN, GAGNEBIN
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Date: 12-11-1

CLG/mc/263629-1
Enclosure

Red-lined claims for the Examiner's convenience:

1. A method for reducing the content of nitrogen oxides in a nitrogen oxide- and oxygen-containing effluent by contacting this effluent in the presence of a deNOx catalyst, optionally in a NOx-storage system, with a reducing gas stream c₁, which comprises CO, H₂ and possibly NH₃ and which stream c₁ has been obtained by converting a stream d₁, which comprises one or more hydrocarbons which are at least partly drawn from a different source than said effluent, with a stream e₁ comprising oxygen and/or water from said effluent stream, if desired in the presence of a reductant forming catalyst, wherein substantially all of the oxygen present in stream e₁ that is used for the formation of stream c₁ is used up.

2. A method according to claim 1, wherein said effluent is the effluent of a step for combusting fuels, which comprises the steps of feeding at least a stream a₁ comprising one or more fuels and a stream b₁ containing excess oxygen with respect to the fuel; wherein said effluent, together with said reducing gas stream c₁ which further optionally comprises one or more hydrocarbons, is contacted with said deNOx catalyst; wherein to stream c₁ no ammonia and no urea is added.

3. A method according to ~~any one of the preceding claims~~ 1, wherein said effluent originates from a combined heat and power unit.

4. A method according to ~~any one of the preceding claims~~ 1, wherein said stream c₁ is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst.

5. A method according to ~~any one of claims~~ 2-4, wherein the fuel in stream a₁ comprises synthesis gas and/or one or more hydrocarbons.

6. A method according to ~~any one of claims~~ 2-5, wherein stream a₁ and stream d₁ comprise at least partly the same compounds.

7. A method according to ~~any one of claims~~ 2-6, wherein the fuel in stream a₁ comprises hydrocarbons which, like the hydrocarbons in stream d₁, have been selected independently from the group consisting of natural gas, methane, diesel oil,

gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, and mixtures thereof.

8. A method according to ~~any one of the preceding claims~~ 1, wherein a reductant forming catalyst is used which is a partial oxidation catalyst for the partial oxidation of hydrocarbons, selected from the group consisting of Pt, Rh, Ru, Pd, Co and Ni and combinations thereof, optionally provided on a support, which support is optionally stabilized with Si, La, Ba and/or Y.

9. A method according to ~~any one of the preceding claims~~ 1, wherein the reductant forming catalyst is a steam reforming catalyst which comprises Ni, Rh and/or Pt.

10. A method according to ~~any one of the preceding claims~~ 1, wherein the reductant forming step is a partial oxidation step, wherein the ratio of hydrocarbons and oxygen is selected such that no complete oxidation takes place.

11. A method according to ~~any one of the preceding claims~~ 1, wherein nitrogen oxides from said effluent are absorbed onto a suitable absorbent and subsequently said reducing gas stream is contacted with said absorbent.

13. A method according to claim 11 ~~or 12~~, wherein the product that is formed in the step wherein stream c~~+~~ is contacted with the structured support is used as fuel for a gas engine.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application : WILLEM HENDRIK BOUWMAN, ET AL.
Application No. : 10/009,622
Filed : December 11, 2001
For : METHOD FOR REMOVING NITROGEN OXIDES FROM
AN OXYGEN-CONTAINING GAS STREAM
Examiner :
Attorney's Docket : VER-154XX

Group Art Unit:

* * * * *

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envelope addressed to: BOX PCT Commissioner for Patents,
Washington, D.C. 20231 on 1-31-02.

By: Charles L. Gagnebin III
Charles L. Gagnebin III
Registration No. 25,467
Attorney for Applicant(s)

* * * * *

SUPPLEMENTAL PRELIMINARY AMENDMENT

BOX PCT
Commissioner for Patents
Washington, D.C. 20231

Sir:

Kindly enter the following Supplemental Preliminary
Amendment in the above-identified application:

In the Claims:

Please cancel Claims 1-23 and insert the following new
claims 24-40 to read as follows:

24. A method for reducing the content of nitrogen oxides in a nitrogen oxide- and oxygen-containing gas stream by contacting this gas stream in the presence of a deNOx catalyst with a reducing gas stream, which comprises CO, H₂ and possibly NH₃ and which reducing gas stream has been obtained by converting hydrocarbons with oxygen and/or water from said nitrogen oxide- and oxygen-containing gas stream, if desired in the presence of a reductant forming catalyst.

25. A method according to claim 24, which substantially all of the oxygen present in the part of said nitrogen oxide- and oxygen-containing gas stream that is used for the reductant formation is used up.

26. A method according to claim 24 wherein said nitrogen oxide- and oxygen-containing gas stream is the effluent of a step for combusting fuels, which comprises the steps of feeding at least a stream a) comprising one or more fuels and a stream b) containing excess oxygen with respect to the fuel; wherein said effluent, together with said reducing gas stream c) which further optionally comprises one or more hydrocarbons, is contacted with said deNOx catalyst; wherein to stream c) no

ammonia and no urea is added; and wherein stream c) has been substantially obtained by subjecting streams d) and e) to a reductant forming step, while stream d) comprises one or more hydrocarbons.

27. A method according to claim 24, wherein said nitrogen oxide- and oxygen-containing gas stream originates from a combined heat and power unit.

28. A method according to claim 24, wherein said reducing gas stream is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst.

29. A method according to claim 26, wherein the fuel in stream a) comprises synthesis gas and/or one or more hydrocarbons.

30. A method according to claim 26, wherein stream a) and stream d) comprise at least partly the same compounds.

31. A method according to claim 26, wherein the fuel in stream a) comprises hydrocarbons which, like the hydrocarbons in stream d), have been selected independently from the group consisting

of natural gas, methane, diesel oil, gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, and mixtures thereof.

32. A method according to claim 24, wherein a reductant forming catalyst is used which is a partial oxidation catalyst for the partial oxidation of hydrocarbons, selected from the group consisting of Pt, Rh, Ru, Pd, Co. and Ni and combinations thereof, optionally provided on a support, which support is optionally stabilized with Si, La, Ba and/or Y.

33. A method according to claim 24, wherein the reductant forming catalyst is a steam reforming catalyst which comprises Ni, Rh and/or Pt.

34. A method according to claim 24, wherein the reductant forming step is a partial oxidation step, wherein the ratio of hydrocarbons and oxygen is selected such that no complete oxidation takes place.

35. A method according to claim 24, wherein nitrogen oxides from the nitrogen oxide- and oxygen-containing gas stream are

absorbed onto a suitable absorbent and subsequently said reducing gas stream is contacted with said absorbent.

36. A method according to claim 35, wherein said absorbent is a barium and/or zeolite comprising alumina washcoat on a structured support.

37. A method according to claim 35, wherein the product that is formed in the step wherein stream c) is contacted with the structured support is used as fuel for a gas engine.

38. A method according to claim 25 wherein

said nitrogen oxide- and oxygen-containing gas stream is the effluent of a step for combusting fuels, which comprises the steps of feeding at least a stream a) comprising one or more fuels and a stream b) containing excess oxygen with respect to the fuel; wherein said effluent, together with said reducing gas stream c) which further optionally comprises one or more hydrocarbons, is contacted with said deNOx catalyst; wherein to stream c) no ammonia and no urea is added; and wherein stream c) has been substantially obtained by subjecting streams d) and e) to a reductant forming step, while stream d) comprises one or more hydrocarbons;

said nitrogen oxide- and oxygen-containing gas stream originates from a combined heat and power unit;

said reducing gas stream is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst;

the fuel in stream a) comprises synthesis gas and/or one or more hydrocarbons;

stream a) and stream d) comprise at least partly the same compounds;

the fuel in stream a) comprises hydrocarbons which, like the hydrocarbons in stream d), have been selected independently from the group consisting of natural gas, methane, diesel oil, gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, and mixtures thereof;

a reductant forming catalyst is used which is a partial oxidation catalyst for the partial oxidation of hydrocarbons, selected from the group consisting of Pt, Rh, Ru, Pd, Co. and Ni and combinations thereof, optionally provided on a support, which support is optionally stabilized with Si, La, Ba and/or Y;

the reductant forming catalyst is a steam reforming catalyst which comprises Ni, Rh and/or Pt;

the reductant forming step is a partial oxidation step;

the ratio of hydrocarbons and oxygen is selected such that no complete oxidation takes place;

nitrogen oxides from the nitrogen oxide- and oxygen-containing gas stream are absorbed onto a suitable absorbent and subsequently said reducing gas stream is contacted with said absorbent.

39. A method according to claim 38, wherein said absorbent is a barium and/or zeolite comprising alumina washcoat on a structured support.


40. A method according to claim 39, wherein the product that is formed in the step wherein stream c) is contacted with the structured support is used as fuel for a gas engine.

REMARKS

The Examiner is encouraged to telephone the undersigned attorney to discuss any matter which would expedite allowance of the present application.

Respectfully submitted,

WILLEM HENDRIK BOUWMAN, ET AL.

By: 
Charles L. Gagnebin III
Registration No. 25,467
Attorney for Applicant(s)

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Boston, MA 02109
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Date: 1-31-2

CLG/mes/266431-1
Enclosure

Title: Method for removing nitrogen oxides from an oxygen-containing gas stream

This invention relates to a catalytic conversion of nitrogen oxides to molecular nitrogen, which nitrogen oxides are formed upon combustion of hydrocarbons and/or synthesis gas (H_2/CO). More particularly, it relates to the conversion of nitrogen oxides in the presence of oxygen such as these are formed, for instance, in the operation of units such as combustion engines under so-called poor or 'lean-burn' conditions, that is: under combustion conditions where an excess of oxygen is present. The invention further relates to the conversion of nitrogen oxides which may be formed in industrial processes, such as nitric acid production.

In the combustion of hydrocarbons with molecular oxygen (for instance originating from air), oxides of nitrogen may be formed due to the prevailing temperature and pressure during the combustion process. These nitrogen oxides, among which NO and NO₂ (usually denoted by NO_x) are very harmful to the environment. Nitrogen oxides are held responsible inter alia for the formation of acid rain and photochemical smog.

Different methods for reducing NO_x emission are known and a number of them are already being applied in practice.

Reducing NO_x emission in stoichiometrically running engines is often accomplished by making use of a so-called three-way catalyst system.

The NO_x conversion catalyst in such systems is capable of converting nitrogen oxides to harmless compounds by reacting them with the reducing combustion products present in the exhaust gas, such as hydrocarbons and CO, to form N₂.

Generally, the known three-way catalysts which effect the reduction of nitrogen oxides are incapable of performing this conversion in the presence of a considerable amount of oxygen.

This is a problem in particular in the removal of nitrogen oxides from the exhaust gases of the above-mentioned lean-burn engines, such as lean-burn gas turbines, diesel engines, gas engines and off-gases of industrial processes, since in such gases, in addition to nitrogen oxides, a considerable amount of oxygen is present. Moreover, hydrocarbons and/or

CO are not present, or present in an insufficient amount, which is a bar to the successful operation of the above-mentioned three-way catalyst systems.

In installations where a considerable amount of oxygen is present in the exhaust gas, therefore, often an amount of reductant is added. The nitrogen oxides can then be converted by the reductant in the presence of a
5 suitable catalyst (deNO_x catalyst). This method is known as the so-called Selective Catalytic Reduction (SCR).

Widely used reductants for the SCR reaction are ammonia and urea. Also, it is known from the literature to use hydrocarbons such as
10 ethylene, propylene and propane as reductant (see, for instance, G.P. Ansell *et al.*, 'Mechanism of the lean NO_x reaction over Cu/ZSM-5', Appl. Catal. B, 2 (1993), pp. 81-100). Other possible reductants are, for instance, CO, H₂ and CH₄, ethanol, hydrocarbons, in particular fuels such as gasoline and diesel oil.

Used most by far as reductant in practical SCR applications, however, are ammonia or urea, or an aqueous urea solution. The use of these agents entails a number of disadvantages. Dosage is extremely critical. If too large an amount of ammonia or urea is charged to the deNO_x catalyst (i.e., more than is necessary to convert the nitrogen oxides), this
15 will lead to so-called ammonia slip. The emission of ammonia from such installations is actually even more harmful from an environmental point of view than the emission of NO_x. Also, it is possible that through oxidation of ammonia, this excess dosage leads to the production of NO_x, which is the very opposite of the object contemplated, viz. reducing the NO_x emission.
20 Another disadvantage of the use of ammonia or urea is the necessary storage thereof and the necessity of periodically supplementing the supply if this is not produced within battery limits. In particular ammonia is very dangerous and noxious and the transport thereof involves great safety and environmental risks. As a consequence of all this, both the investment costs
25 and the operational costs of this technique are high.
30

Although the choice of other reducing agents, such as, for instance, the above-mentioned hydrocarbons, might partly solve these problems, there still remain disadvantages, such as the necessity of separate transport and storage. The attendant safety and environmental hazards are often
35 unacceptable.

This problem would be solved by using as a reducing agent the same fuel that is used or is present in the engine, the (gas) turbine or the combustion process in industrial processes. The hydrocarbons which are present in, for instance, diesel oil and gasoline, however, prove not to be sufficiently active to convert NOx under process conditions with an acceptable rate and selectivity.

It is known to start from an SCR catalyst, with the required reducing agent being manufactured from an available source of hydrocarbons, such as methanol, LPG and natural gas, optionally under addition of H₂ which has been obtained from electrolysis or which is stored in storage tanks.

In DE-A-44 04 617 a technique is described whereby, using an electrically heated reactor, a hydrocarbon-containing fuel is catalytically cracked at 200 to 700°C and the cracking products are further activated with air before they are added as reducing agent to the exhaust gas, whereupon the total gas stream is passed over an SCR catalyst.

According to DE-A-196 00 558, also cracked hydrocarbons are used as reducing agent. These hydrocarbons have been obtained from diesel. According to this known method, under SCR conditions, hydrogen is added to the exhaust gas to reduce NOx over the SCR catalyst where the cracked hydrocarbons are not sufficiently active. The hydrogen originates from a storage tank or is made through electrolysis or methanol reforming.

The addition of hydrogen as reducing agent for the reduction of NOx in the exhaust gases of continuous combustion processes is described in DE-A-42 30 408. There, too, the hydrogen can be obtained by electrolysis or from reforming through steam reforming or partial oxidation (PO) of hydrocarbon containing fuel. To obtain a hydrogen stream with as little carbon monoxide as possible, two shift reactors are placed behind the reformer to convert the CO for the most part with water vapor to form hydrogen and CO₂.

The use of in situ produced hydrogen as a reducing agent for the reduction of NOx from exhaust gas of internal combustion engines is described in EP-A-0 537 968. In this document, the technique of reforming (steam reforming and partial oxidation) of hydrocarbon containing fuels is described. According to this publication, the conditions must be chosen such

that the amount of CO in the hydrogen is so low that the CO concentration cannot yield any problems regarding emissions. The temperature needed for reforming is achieved by making use of the heat of the exhaust gases. The reduction of NOx is carried out over an SCR catalyst. As oxidizing agent for the partial oxidation, air is mentioned.

According to the present invention, in a reductant-forming step, hydrocarbons are converted to a reductant stream under suitable conditions, optionally by contacting them with a reductant forming catalyst. The hydrocarbons in the product stream of the reductant forming step can be unreacted hydrocarbons from the feedstock of this step, but may also be smaller hydrocarbons which have been formed by cracking reactions during the reductant forming step.

The reductants can be prepared, by the use of a reductant forming catalyst, from, for instance, residues of hydrocarbons which are contained in the exhaust gas of the unit in which the combustion takes place. It is also possible to draw these hydrocarbons from a different source, for instance the fuel for the combustion unit, which is already available in situ. Combinations of effluent and such a different source are naturally also possible. The oxygen needed for this step at least partly originates from the off-gas to be treated, i.e., the nitrogen oxide- and oxygen-containing gas. Preferably, substantially all oxygen present in the part of the off-gas used for the reductant formation is used for forming the reducing gas stream. The hydrocarbons present in this portion of the off-gas can also be converted to reducing compounds in this step. The hydrocarbons present in the off-gas can then also be converted to reducing compounds in this step.

Through the use of a suitable reductant forming process, optionally utilizing a catalyst, the reductants necessary for the catalytic reduction of NOx, in the form of CO and/or H₂, optionally supplemented with hydrocarbons, can be prepared from hydrocarbons in situ, so that the above-outlined drawbacks in removing NOx under oxygen-rich conditions can be eliminated at least partly.

In addition to the above-mentioned reductants, CO and/or H₂, optionally supplemented with hydrocarbons, also ammonia (NH₃) can be formed in the presence of hydrogen and nitrogen, under suitable process

conditions, because of the position of the chemical equilibrium
 $3\text{H}_2 + \text{N}_2 = 2\text{NH}_3$. As mentioned above, ammonia is a good reductant.

The use of a part of the exhaust gases as a source of hydrocarbons and certainly as an oxidation source for the (catalytic) partial oxidation and/or steam reforming has as a major advantage over the existing technology that the emission of uncombusted hydrocarbons can be minimized, the oxygen content in the total exhaust gas stream decreases, so that the conditions for deNO_x-ing improve and the energy present in the form of heat in the exhaust gases can be used directly for the catalytic process without making use of extra heat exchanging surface.

Another advantage is that no pure H₂ or H₂ with small amounts of carbon monoxide needs to be produced. As a consequence, it is possible to omit shift reactors and membrane technology.

The reductant forming step can be a partial oxidation step, in which use can be made of a partial oxidation catalyst. In addition, it is possible to carry out the partial oxidation without catalyst, for instance by supplying a controlled amount of energy to a fuel stream, for instance by means of an electrical discharge. Also, the reductant forming step can be a steam reforming step in which use is made of a steam reforming catalyst. A combination of partial oxidation and steam reforming is also possible.

A suitable reductant forming catalyst is, for instance, a partial oxidation catalyst. In the presence of such a catalyst, the partial oxidation of hydrocarbons takes place. As mentioned, according to the invention, the oxygen needed for this partial oxidation is derived from the effluent of the combustion unit, optionally supplemented with oxygen obtained from elsewhere, coming, for instance, from added air. The product stream of the partial oxidation step is highly suitable for use as reductant stream.

Another possibility of preparing a stream comprising H₂ and/or CO, and optionally hydrocarbons, from a stream comprising hydrocarbons, is the use of a so-called steam reforming. In steam reforming, in addition to hydrocarbons, water is to be added to the steam reforming step. This water may originate from the effluent of the combustion engine, from a separate stock, or from a combination of these two sources. In steam reforming, hydrocarbons are converted with water (steam) to a mixture of hydrocarbons, such as methane, and/or H₂ and CO₂. As a consequence of

chemical equilibria, in addition to these components, CO may also be present. The mixture thus formed is very suitable to be used as a reductant stream.

5 Next, the reductant stream, together with the effluent of the combustion unit, is contacted with a deNOx catalyst so that the desired conversion of nitrogen oxides takes place.

10 The in situ production of the reductant offers a number of important advantages. Thus, according to the invention, the supply of reductants can take place continuously, in the case of natural gas, or in any case simultaneously with the supply of the fuel for the combustion unit, and it is no longer necessary to have and maintain a separate stock thereof. This can be practical, for instance, when used in mobile combustion units, such as trucks or passenger cars, because then no separate storage tanks for the reducing agent need to be present. This may also be an important
15 advantage in stationary units. The fact that no ammonia and urea (whether or not as a urea solution) are used, at least do not need to be drawn from elsewhere, is also an advantage because the above-outlined disadvantages associated with the use of these reductants no longer occur.

20 Another advantage of the invention arises if hydrocarbons from the effluent of the combustion unit are used as feedstock for the reductant forming step, as a result of which the amounts of hydrocarbons in the exhaust gas will then be lowered in that they are used for the reaction with NOx. Such a reduction is favorable, since the emission of hydrocarbons is undesirable from an environmental point of view. Another advantage of the
25 use of the effluent of the combustion unit for the formation of the reducing gas is the decrease of the amount of hydrocarbons needed for the reductant formation.

30 Accordingly, the present invention is characterized by a method for reducing the content of nitrogen oxides in a nitrogen oxide- and oxygen-containing gas stream by contacting this gas stream in the presence of a deNOx catalyst with a reducing gas stream, which comprises CO, H₂ and possibly NH₃ and which reducing gas stream has been obtained by converting hydrocarbons with the oxygen from the nitrogen oxide- and oxygen-containing gas stream, optionally in the presence of a reductant
35 forming catalyst.

According to a preferred embodiment, the nitrogen oxide- and oxygen-containing gas stream is the effluent of a step for combusting fuels, which comprises the steps of feeding at least a stream a) comprising one or more fuels and a stream b) containing excess oxygen with respect to the fuel, which further comprises nitrogen, wherein the effluent, together with the reducing gas stream c) which further optionally comprises one or more hydrocarbons, is contacted with the deNO_x catalyst, wherein to stream c) no ammonia and no urea is added and wherein stream c) has been substantially obtained by contacting streams d) and e) with the reductant forming catalyst, stream d) comprising one or more hydrocarbons and stream e) comprising oxygen and water.

The combustion unit is suitable for generating heat, and optionally energy. The combustion unit can work on the basis of a flame, but the combustion in the combustion unit can also proceed by catalytic route. Preferably, the combustion unit is a gas engine, gas turbine, diesel engine or gasoline engine.

A stream c) which is substantially free from externally added ammonia and urea is understood to mean that to this stream, in accordance with the invention, no reductants of this type need to be added. Still, ammonia and possibly compounds derived therefrom, such as urea, can be present as a result of the above-mentioned equilibrium reaction of N₂ and H₂.

According to the present invention, the NO_x is reduced with a mixture of H₂, CO and possibly NH₃. In addition, hydrocarbons which have not been (entirely) converted can be present in the reducing gas stream. These hydrocarbons also work as reductants. The reducing gas mixture is obtained by converting the hydrocarbons from a part of the exhaust gas stream with optionally extra added hydrocarbons, by means of (catalytic) partial oxidation, steam reforming or a combination of both technologies by means of the oxygen and water vapor present in the same part of the exhaust gas stream and optionally externally added air and/or water vapor. Optionally, extra reducing agent such as hydrogen can be added to the reducing gas mixture obtained.

Particularly preferred is the method and the apparatus suitable therefor according to the invention, wherein the engine comprises a heat

exchanger, such that at least a portion of the heat released upon the combustion can be usefully employed, for instance for heating, as of greenhouses or other spaces. Such units, in which at the same time both heat and energy are generated, the energy being typically in the form of electrical power, are also referred to as combined heat and power units or total energy plants.

The invention can also be applied in various means of transport such as vessels, airplanes, trucks and passenger cars and trains provided with a hydrocarbon-fired locomotive engine.

The method according to the invention and the apparatuses suitable therefor is suitable in particular for operating combustion units under so-called lean-burn conditions, i.e., conditions where the ratio between the streams a) and b) is selected such that the amount of oxygen is at least the amount needed for the complete combustion of the fuels in stream a). These are the conditions under which oxygen is present in the effluent of the combustion unit and wherein, with advantage, the deNO_x reaction with the effluent of the catalytic partial oxidation step can be carried out.

Suitable fuels for the feedstock a) are hydrocarbons and/or synthesis gas (CO/H₂ mixture).

Preferably, the hydrocarbons for the reductant forming step are at least partly drawn from the same source as the fuel for the combustion unit. In this case, stream a) and stream d) comprise the same components.

In order to reduce the content of hydrocarbons in the emission of the apparatus for the method according to the invention, the hydrocarbons that are present in the effluent of the combustion unit are at least partly used as feedstock for the reductant forming step, which may or may not be supplemented with a hydrocarbon stream drawn from elsewhere.

In order to keep the oxygen content low when contacting the streams with the deNO_x catalyst, the effluent of the combustion unit is used as oxygen source, which may or may not be supplemented with an oxygen stream drawn from elsewhere.

As fuel for the combustion unit and/or as feedstock for the reductant forming step, besides synthesis gas, in principle all hydrocarbons suitable therefor can be used. It is practically preferred, however, when the

hydrocarbons from stream a) and d) are selected independently from the group consisting of natural gas (which comprises substantially methane), methane, diesel oil, gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, derivatives and mixtures thereof.

5 The catalyst for converting nitrogen oxides can be selected from the group of catalysts which catalyze the reduction of NO_x, such as the conventional catalysts for removal of NO_x. Preferably, these are selected from the group comprising zeolites, metal-exchanged zeolites, such as Co-, Cu- and/or Ce-exchanged zeolites, Pt, Rh and/or Ir catalyst, optionally
10 provided on a support such as a washcoat which may further comprise Ba, La, Y, Sr, Pr, Ce, Si, Ti, Al and/or Zr.

 The catalyst for the partial oxidation of hydrocarbons can be selected from the group consisting of Pt, Rh, Ru, Pd, Co and Ni, if desired provided on suitable supports such as Al₂O₃, SiO₂, TiO₂, ZrO₂,
15 silica/alumina-zeolites and mixtures thereof, optionally stabilized with, for instance, Si, La, Ba or Y and mixtures thereof.

 The steam reforming catalyst which is capable of converting a mixture of hydrocarbons and water to a mixture of H₂, CO, CO₂ and/or hydrocarbons can be any conventional steam reforming catalyst, which may
20 or may not be supported, according to conventional techniques, as known to the skilled person. Preferably, the steam reforming catalyst is a supported catalyst comprising Ni, Rh and/or Pt.

 For operating an apparatus according to the invention, factors such as hydrocarbon/oxygen ratio, temperature, pressure, residence time and/or
25 amount of catalyst for the partial oxidation of hydrocarbons should be selected such that no complete oxidation takes place. The molar hydrocarbon/oxygen ratio is expressed in λ_{PO} , such that for stoichiometric ratios (i.e., precisely sufficient oxygen to effect the complete combustion of the fuel), $\lambda_{PO} = 1$. According to the invention, $\lambda_{PO} < 1$. Preferably, $0.2 < \lambda_{PO} <$
30 0.7 . λ_{PO} is controllable by adjusting the air/fuel amount and is dependent on the hydrocarbons used.

 The temperature for the reductant forming step is generally between 250 and 1100°C. The residence time for the reductant forming step is generally between 200 en 150,000 h⁻¹. Although the pressure will also
35 have an influence, it is generally dictated by the other process conditions. In

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general, the pressure will be atmospheric or slightly higher and not higher than 50 bars.

The use of an SCR catalyst as a method of converting NO_x with a reducing agent to N₂ often has the limitation that sufficient conversion of NO_x can be achieved in a limited temperature frame. Thus, in DE-A-196 00 558, as a practical example, a conversion curve of NO_x as a function of the temperature is given, in which a conversion of 40% is achieved. This is typical of SCR systems that do not work with NH₃ or urea as reducing agent. Selecting the process conditions of the reductant forming step such that also NH₃ is generated is therefore advantageous because then a higher NO_x conversion can be achieved.

When a high conversion of NO_x is required, as, for instance, in the application of exhaust gases as fertilizing gas in horticulture, it is better for the method according to the present invention to be carried out with an NO_x storage system, instead of an SCR system. If the deNO_x step is carried out with such an NO_x storage system (also referred to as NO_x Storage and Reduction Catalyst, NSR), such as described, for instance, in N. Takhashi et al., Environmental Catalysis, p. 45, (1995), a very high conversion in NO_x can be obtained. According to this method, nitrogen oxides are absorbed from the nitrogen oxide- and oxygen-containing gas stream to a suitable absorbent and subsequently, for instance by switching, the reducing gas stream is contacted with the absorbent. The deNO_x step is thus operated discontinuously. A very effective NO_x removal can thus be obtained.

With this NO_x storage system, it is possible to absorb NO_x in an oxidizing medium ($\lambda > 1$), and in a reducing medium ($\lambda < 1$) to convert both the NO_x in the exhaust gas and the adsorbed NO_x to nitrogen. The catalyst in the NO_x storage system can very suitably consist of platinum provided on a barium-containing and/or zeolite-comprising alumina washcoat. The barium present can react with NO_x to barium nitrate. This nitrate salt decomposes in a reducing medium to barium and N₂.

The NO_x storage system can be operated according to the invention by passing the exhaust gases through the NO_x storage system until the system is saturated with NO_x. Thereafter, regeneration can be done with a reducing agent which has been obtained as described above. Optionally, this

reducing agent can be supplemented with a reduction agent drawn from elsewhere.

Preferably, this NO_x storage system is designed with minimally two parallel beds. One bed is used to absorb NO_x, while the other bed is regenerated. As soon as the former bed is saturated and/or the second bed is sufficiently regenerated, the streams are switched, so that the regenerated bed can proceed to absorb NO_x and the loaded NO_x bed is regenerated in that the absorbed NO_x is converted to nitrogen.

The effluent of the regeneration step of the NO_x storage bed can be recirculated with advantage and, together with the inlet air, be passed to the inlet of the combustion unit (for instance a gas engine). This provides at least two advantages. In the first place, in this way no CO-containing gas needs to be discharged. In the second place, according to this embodiment, it is of less importance to obtain a complete conversion of hydrocarbons to CO/H₂ in the preceding preparation step of the reducing gas stream.

Both the deNO_x catalyst and the reductant forming catalyst can be present in the method according to the invention in the forms known to those skilled in the art, as in the form of a bed of granules, extrusions, granules, and/or pellets, or provided on ceramic or so-called metal monoliths, or differently structured forms.

The use of the catalyst in structured form is preferred because this allows other relevant factors for the method, such as pressure drop, mixing, contact time, heat management, mechanical strength and life, to be tailored to the prevailing conditions through suitable choices, and the method can thereby be optimized.

The invention is therefore characterized by the use of a catalyst suitable for converting nitrogen oxides in combination with either a catalyst suitable for the partial oxidation of hydrocarbons, or a catalyst suitable for steam reforming, for converting nitrogen oxides in generating heat and optionally energy from hydrocarbons, without this requiring ammonia or urea to be added externally.

Amended claims

1. A method for reducing the content of nitrogen oxides in a nitrogen oxide- and oxygen-containing effluent by contacting this effluent in the presence of a deNO_x catalyst, optionally in a NO_x-storage system, with a reducing gas stream c), which comprises CO, H₂ and possibly NH₃ and which stream c) has been obtained by converting a stream d), which comprises one or more hydrocarbons which are at least partly drawn from a different source than said effluent, with a stream e) comprising oxygen and/or water from said effluent stream, if desired in the presence of a reductant forming catalyst, wherein substantially all of the oxygen present in stream e) that is used for the formation of stream c) is used up.
2. A method according to claim 1, wherein said effluent is the effluent of a step for combusting fuels, which comprises the steps of feeding at least a stream a) comprising one or more fuels and a stream b) containing excess oxygen with respect to the fuel; wherein said effluent, together with said reducing gas stream c) which further optionally comprises one or more hydrocarbons, is contacted with said deNO_x catalyst; wherein to stream c) no ammonia and no urea is added.
3. A method according to any one of the preceding claims, wherein said effluent originates from a combined heat and power unit.
4. A method according to any one of the preceding claims, wherein said stream c) is obtained by carrying out a partial oxidation step, utilizing a partial oxidation catalyst, or a steam reforming step, utilizing a steam reforming catalyst.
5. A method according to any one of claims 2-4, wherein the fuel in stream a) comprises synthesis gas and/or one or more hydrocarbons.
6. A method according to any one of claims 2-5, wherein stream a) and stream d) comprise at least partly the same compounds.

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7. A method according to any one of claims 2-6, wherein the fuel in stream a) comprises hydrocarbons which, like the hydrocarbons in stream d), have been selected independently from the group consisting of natural gas, methane, diesel oil, gasoline, fuel oil, methanol, ethanol, naphtha, kerosene, ethane, propane, butane, LPG, and mixtures thereof.

8. A method according to any one of the preceding claims, wherein a reductant forming catalyst is used which is a partial oxidation catalyst for the partial oxidation of hydrocarbons, selected from the group consisting of Pt, Rh, Ru, Pd, Co and Ni and combinations thereof, optionally provided on a support, which support is optionally stabilized with Si, La, Ba and/or Y.

9. A method according to any one of the preceding claims, wherein the reductant forming catalyst is a steam reforming catalyst which comprises Ni, Rh and/or Pt.

10. A method according to any one of the preceding claims, wherein the reductant forming step is a partial oxidation step, wherein the ratio of hydrocarbons and oxygen is selected such that no complete oxidation takes place.

11. A method according to any one of the preceding claims, wherein nitrogen oxides from said effluent are absorbed onto a suitable absorbent and subsequently said reducing gas stream is contacted with said absorbent.

12. A method according to claim 11, wherein said absorbent is a barium and/or zeolite comprising alumina washcoat on a structured support.

13. A method according to claim 11 or 12, wherein the product that is formed in the step wherein stream c) is contacted with the structured support is used as fuel for a gas engine.

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(54) Title: METHOD FOR REMOVING NITROGEN OXIDES FROM AN OXYGEN-CONTAINING GAS STREAM

(57) Abstract: A process for removing nitrogen oxides from a gas stream containing nitrogen oxides and oxygen involves bringing the gas stream into contact with a reducing gas stream in the presence of a deNO_x catalyst. The reducing gas stream contains CO, H₂ and, optionally, NH₃; it is produced by reacting hydrocarbons with the oxygen from the above mentioned nitrogen oxide- and oxygen-containing gas stream. A catalyst, such as an oxidising or stream-reforming catalyst may be used to promote the reaction which produces the reducing gas stream.

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Declaration and Power of Attorney Patent Application (Design or Utility)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: "Method for removing nitrogen oxides from an oxygen-containing gas stream."

the specification of which

- ☐ is attached hereto
☒ was filed on December 11, 2001 ✓ as application serial no. 10/009,622 ✓
 and or PCT International Application number PCT/NL00/00404 and was amended
 on (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the U.S. Patent and Trademark Office all information know to me to be material to patentability as defined in 37 C.F.R. §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or 35 U.S.C. §365(b) of any foreign application(s) for patent or inventor's certificate, or 35 U.S.C. §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate of PCT International application having a filing date before that of the application on which priority is claimed.

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I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below:

Prior Provisional Application(s)	
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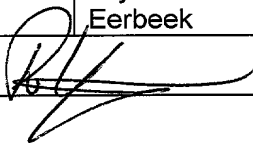
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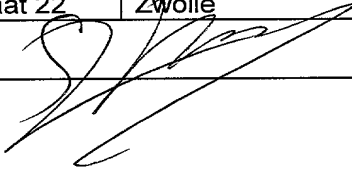
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